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Original Research Article

Spatial and temporal analysis of ambient carbonyls in a densely populated basin area of central Taiwan

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ABSTRACT

The spatial and temporal distributions of ambient carbonyls during the southwest and northeast monsoon seasons in a densely populated basin area were analyzed. The results show that the major carbonyls in ambient were ketones during the both monsoon seasons. The concentrations of carbonyls at daytime and nighttime were in the order: urban > rural > suburban and urban > suburban > rural areas, respectively. The higher carbonyl concentrations occurred in urban area all day, but maximum variation of carbonyl concentrations occurred in rural area. The orders of carbonyl concentrations at daytime and nighttime were irregular during the northeast monsoon season; the variations of carbonyl concentrations in the area were all low. The effects of urban heat island phenomena on the area were negligible, especially during the northeast monsoon season. The ranges of average ratio (northeast to southwest monsoon seasons) of total carbonyl concentrations in urban, suburban, rural areas were 1.0–1.6, 1.5–3.5, and 1.8–2.4, respectively. Because the clear results were obtained during the both monsoons that ambient carbonyls increased with increasing volatile organic compound (VOC) concentrations, the simulation results of VOCs by CMB8.2 model were used to understand the contribution of carbonyls from the primary sources. The order of VOCs and carbonyls source contributions were vehicle exhaust > hi-tech industry > traditional industry > ship and gasoline stations during the both monsoon seasons. Therefore, in order to effectively reduce the carbonyls in ambient air, pollution from vehicle and hi-tech industry must be managed and controlled.

1. Introduction

Carbonyl compounds in ambient air are well known for their adverse effects on human health, especially on eye and lung irritations [1,2]. A large amount of data has implicated reactive carbonyls as neurotoxic mediators of oxidative damage in the progression of Alzheimer's disease and other neurodegenerative diseases [3]. Exposure of red blood cells to carbonyl compounds leading to Maillard reaction. It caused a marked decrease in red blood cell deformability [4]. Several studies have indicated that carbonyl emissions from cook stoves when the combustion is incomplete [5,6]. Kabir et al. [7] quantified the emission concentrations of a number of carbonyl compounds during the combustion of commonly used barbecue charcoal. Kim et al. [8] quantified the emission concentrations of 13 carbonyl compounds released directly from various anthropogenic source processes in a large industrial complex. Researchers have also identified motor vehicles as the leading sources of carbonyls emission in non-industrial area [9–11].

The gas-phase reaction of ozone with alkenes is of critical importance in atmospheric chemistry. It leads several products including carbonyls [12]. Most secondary production of formaldehyde is expected to occur during the atmospheric oxidation of alkenes. Additionally, it is formed more slowly from both the oxidation of alkanes and aromatic compounds [13,14].

The topography of Taichung area (about 2500 km^2) is complex, which includes coast, tablelands, basin, hills, and mountains (Fig. 1). At the end of 2012, approximately 2.3 million and 0.9 million peoples lived in the basin and coastal areas, respectively. A densely populated area is located within the basin center, with surrounded by rural area. Additionally, topographical and meteorological factors, such as basin and monsoons, can drastically affect

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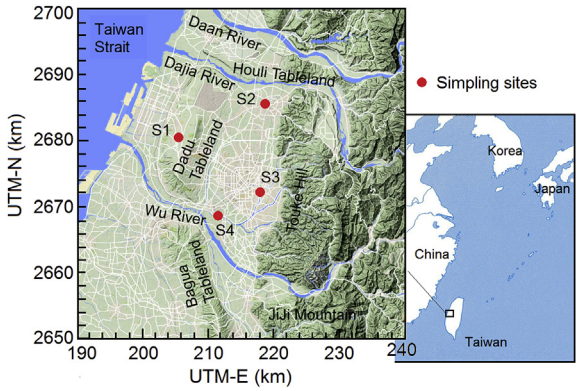

Fig. 1. Location of sampling sites in Taichung Basin. The Taichung Basin faces south toward the Wu River, north toward the Dajia River, East toward the Touke Hill, West toward the Dadu Tableland, Southwest toward the Bagua Tableland, and Southeast toward the JiJi Mountain.

levels of carbonyls and volatile organic compounds (VOCs) in ambient air of the area. The weather conditions in the area are different. For example, urban heat island effect occurs in the basin area; the clear monsoon effect tale places in the coastal area. Also, a city with a million people would cause a 4.4 °C warming [15]; urban heat island effect in this basin with intensive and frequent occurrence resulting in more pollutant accumulation in the basin. Therefore, characterizing the variation of ambient air pollutants, especially carbonyls and VOCs, with sources, monsoons, and terrain in the target area is crucial.

In some previous studies, the measurement of ambient carbonyls based on 2,4-dinitrophenylhydrazine (DNPH) or O-(2,3,4,5,6-pentafluorobenzyl)hydroxylamine (PFBHA) derivatisations is utilized. It is followed by separation and detection of the hydrazones with high-performance liquid chromatography/UV–Vis absorption (HPLC/UV-V) or gas chromatography/mass spectrometry (GC/MS) [16–19]. The procedures of these methods are complicated and these methods are unable to measure ambient VOCs at the same time. In present study, US EPA TO-15 Method was employed to measure ambient carbonyls and VOCs. This method determines VOCs in air collected in specially-prepared canisters, analyzed by GC/MS, which applies to ambient concentrations of VOCs above 0.5 ppbv and typically requires VOC enrichment by concentrating up to one liter of a sample volume. The sample air was collected and stored in canisters with subsequent GC/MS analysis. It is due to the fact that α,β-unsaturated carbonyls are unstable when collected in electropolished canisters [20]. Therefore in this study, some specially prepared (without humidification) silcosteel canisters with air sampling kites were selected for collecting ambient air samples to avoid this problem.

In previous studies, several approaches have been applied for understanding the source apportionment of carbonyls. The multilinear regression (MLR) method [21,22] and positive matrix factorization (PMF) model [23] are two commonly used techniques. The MLR method separates primary and secondary carbonyls based on their correlations with markers for primary emission (e.g., CO, ethyne) and secondary production (e.g., O_3, glyoxal). The PMF technique is used to derive a set of source composition profiles. Each identifies a mix of compounds associated with a particular category of emissions (e.g., on-road emissions, solvent manufacturing) [24]. However, the MLR method heavily depends on the selection of markers, and improper markers may bias source apportionment results [23]. The uncertainty from rotational ambiguity in the PMF solutions could affect the species loadings in the source profile matrix [25]. Thus, this study uses bimestrial field measurements and the chemical mass balance model (CMB8.2) to analyze VOC emissions and further understanding of carbonyls emissions from various possible sources in the area. For the simulation of CMB8.2 model, 10 source profiles of VOC emissions were established in this study.

2. Materials and methods

2.1. Field measurements

Fig. 1 shows the map of Taichung Basin with sampling sites. Taichung Basin faces south toward the Wu River; north toward the Dajia River; east toward the Touke Hill; west toward the Dadu Tableland; southwest toward the Bagua Tableland; and southeast toward the JiJi Mountain. Four sampling sites were selected to measure the ambient concentration of carbonyls. Sampling site S1 was chosen for its location outside the basin. In the basin, the sampling sites S2 and S4 were selected to follow the pollutant transport in which there were two passes channeling air masses in the basin. Sampling site S3 was selected because of its proximity to a prevailing strong heat island effect at the center of basin [26,27].

Ho et al. [28] pointed out that no photochemical reactions occur between 06:00 and 09:00 (early morning); the period of less photochemical reactions between 09:00 and 12:00 (morning); the ground-level atmosphere mixes rapidly increasing photochemical reactions, causing VOC fraction variations between 12:00 and 15:00 (noon); the later period of photochemical reactions between 15:00 and 18:00 (afternoon); and the period after photochemical reaction between 18:00 and 21:00 (evening). In this study, bimestrial samples from May 2011 to February 2012 were collected simultaneously at sites S1–S4 for 3 d (72 h). The samples were taken four times per day for the periods of 4 h: 08:00–12:00 (morning), 12:00–16:00 (afternoon), 16:00–20:00 (evening), and 20:00–24:00 (night).

The ambient air samples at four sampling sites (S1–S4) were simultaneously collected following the US EPA Compendium Method TO-15. A specially prepared (without humidification) in vacuum silcosteel canisters (RESTEK SilcoCan cat. no. 24114) was used with air sampling kites (RESTEK Veriflo SC423XL flow controllers). After sampling, the ambient carbonyls and VOC samples were analyzed by GC/MS (Shimadzu QP-5050A) with thermal-adsorption equipment (ENTECH 7100A Preconcentrator) according to US EPA Method TO-15. A 0.25 mm Chrompack DB-1 60 m-length capillary column was employed in the GC/MS. 102 standard curves spectrum gases were calibrated using the standard gases in U.S. EPA Method TO-14A calibration mix (RESTEK cat. no. 34400), Method TO-15 ozone precursor mixtures (RESTEK cat. no. 34420), and prepared standard gases. The detection limits of VOC concentrations were in the μg m^{-3} range.

The equipment blanks were analyzed in a laboratory with high purity nitrogen. The results were all lower than 2 times of the method detection limit. Because these standard curves did not contain all compounds in ambient air, a method of relative response factor (RRF) was used to quantitatively determine compounds for each non-standard curve in the air sample. In addition, the experiments were conducted on 102 standard gases. Also, a number of pure statistical RRF equations were proposed to estimate the concentrations of other alkanes, alkenes, aromatics, carbonyls, alcohols, and chlorohydrocarbons. These equations depend on a number of the VOC properties. The most important of which are the molecular weight, number of double bond, boiling point, number of branched chain, number of carbon, number of chlorine, stereo structure, and positions of function group. These RRF equations are as follows:

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i. Alkanes, $R^2 = 0.994$

$$RRF_{\text{paraffin/heptane}} = -0.388 - 0.096Mw + 1.43Nc - 0.003Nb - 0.341Nr + 0.003Bp \quad (1)$$

ii. Alkenes, $R^2 = 0.856$

$$RRF_{\text{olefin/hexene}} = 2.979 + 0.094Mw + 0.169Nd + 0.251Si - 0.028Bp \quad (2)$$

iii. Aromatics, $R^2 = 0.865$

$$RRF_{\text{aromatic/toulene}} = -1.008 + 0.041Mw - 1.016Nc + 0.051Nb - 0.148Pf + 0.898Nd + 0.014Bp \quad (3)$$

iv. Carbonyls, $R^2 = 0.707$

$$RRF_{\text{carbonyl/acetaldehyde}} = -13.295 - 0.116Mw + 1.065Nb + 0.066Bp \quad (4)$$

v. Alcohols, $R^2 = 0.772$

$$RRF_{\text{alcohol/1-butanol}} = -4.895 - 0.548Nc + 0.025Nb + 0.021Bp \quad (5)$$

vi. Chlorohydrocarbons, $R^2 = 0.952$

$$RRF_{\text{chlorohydrocarbon/tri chloroethane}} = -3.015 - 0.440Nc - 0.419Ncl - 0.313Nb + 0.375Nd + 0.016Bp \quad (6)$$

where $RRF_{i/s}$ is the relative response of the instrument detector to i target analyte compared to s standard; Mw is molecular weight (g mol^{-1}); Nd is number of double bond (C=C); Bp is boiling point (K); Nr is cyclohydrocarbon (0 = no, 1 = yes); Nb is number of branched chain; Nc is number of carbon; Ncl is number of chlorine atoms; Si is stereo structure of isomer (1 = tran, 2 = cis); and Pf is positions of function groups (1 = meta, 2 = para, 3 = ortho).

The quantitative equation of a target analyte is as follows:

$$C_i = \frac{A_i}{A_s} \cdot \frac{C_s}{RRF_{i/s}} \quad (7)$$

where C_i and A_i are the concentration and peak area of i target analyte, respectively; C_s and A_s are the concentration and peak area of the corresponding s standard, respectively.

2.2. Chemical mass balance model

In this study, CMB8.2 model is used to identify the sources and relative ambient VOC contributions by selecting source profiles and fitting species. It assists to understand the compositions and source reconciliations in Taichung basin in detail. Multiple linear least squares regression is used in CMB8.2 model to assess pollutant contributions from various sources based on observation data at sampling sites. In our previous studies, the applications of CMB8.2 modelling were successful and accurate to characterize the effects of terrain and monsoon on ambient pollutants; the VOCs from control device exhaust in a science park; and the benefits for improved air quality [29–32]. In this study the performance of the CMB8.2 model is evaluated by using several statistical indices, including R-squared ($R^2 \geq 0.8$), Chi-squared ($\chi^2 \leq 4.0$), and percentage mass (ratio = 80–120%). The CMB-8.2 model is based on the following equation:

$$C_{ij} = \sum_{k=1}^{N} m_{ik} S_{kj}, \quad i = 1, 2, \ldots, m \text{ and } j = 1, 2, \ldots n \quad (8)$$

where C_{ij} is the measured concentration of species i in sample j, m is the total number of chemical species used for CMB8.2 calculation, m_{ik} is the mass fraction of species i in source k, S_{kj} is the total mass contributed from source k and in sample j, n is the number of samples, and N is the number of emission sources.

2.3. VOCs sources

Fig. 2a shows the bubble plot of VOC emissions from stationary sources, which are mainly from photovoltaic industry (PVI), semiconductor manufacturing (SCM), printer (PR), plastic (PL), surface coating (SC), and polyurethane synthetic leather (PU) processes, in the study area. Data of stationary emissions were obtained from the TEDS7.1 database published by the Environmental Protection Administration, Taiwan (http://www.ctci. Com.tw/air-ei/new_main2-1.htm). From Fig. 2a, it can be found that most VOC emission sources were located in the basin area, especially S3. As mentioned previously, the population in the basin area (urban and suburban) was higher than those in coastal (rural). The population densities in the urban (S3), suburban (S2 and S4), and rural (S1) areas were approximately 3730, 1740, and 1300 person km^{-2}, respectively.

Main road location in the Taichung area is shown in Fig. 2b. The amount of VOCs emitted from vehicle exhausts was estimated for each grid using the following equation:

$$E = f_i \sum_{l=1}^{n} L_l P_l T_l \quad (9)$$

where E is the VOC emission amount (kg yr^{-1}); L_l is the road length l (km); P_l and T_l are the passenger cars (PCU h^{-1}) and traveling time (h) on the roads l per year; f_i is the vehicle exhaust VOC emission factors (kg km^{-1} PCU^{-1}); and n is the total number of roads. The vehicle emission factors f_i, and traffic data P_l were obtained from data on the websites of the EPA (http://www.ctci.com.tw/air-ei/new_main2–1.htm) and Ministry of Communications (http://www.thb.gov.tw/index.aspx), Taiwan, respectively. The amount of VOCs emitted from vehicle exhausts in the urban (S3), suburban (S2 and S4), and rural (S1) were approximately 2500, 1640, and 2080 ton yr^{-1}, respectively.

3. Results and discussion

3.1. Field measurement results

In Taiwan, the major monsoon seasons consist of the southwest monsoon (May–September) and northeast monsoon (October–April). Figs. 3 and 4 show the wind roses for the (a) southwest and (b) northeast monsoons during the sampling periods, respectively. Each wind rose was drawn using simultaneously collected meteorological data at S1, S2, S3, or S4 site for 72 h, from which the general wind patterns were obtained for two monsoons (Figs. 3 and 4).

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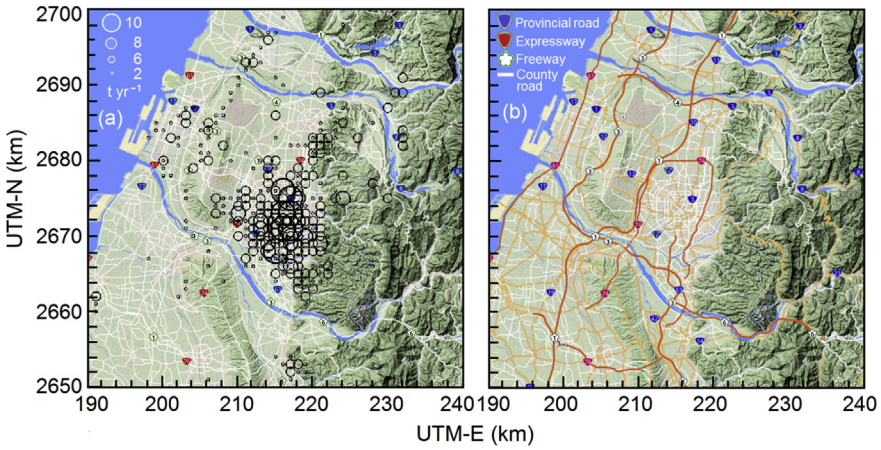

Fig. 2. Bubble plot of VOC emission stationary sources (a) and schematic diagram of main road location (b) in the Taichung area.

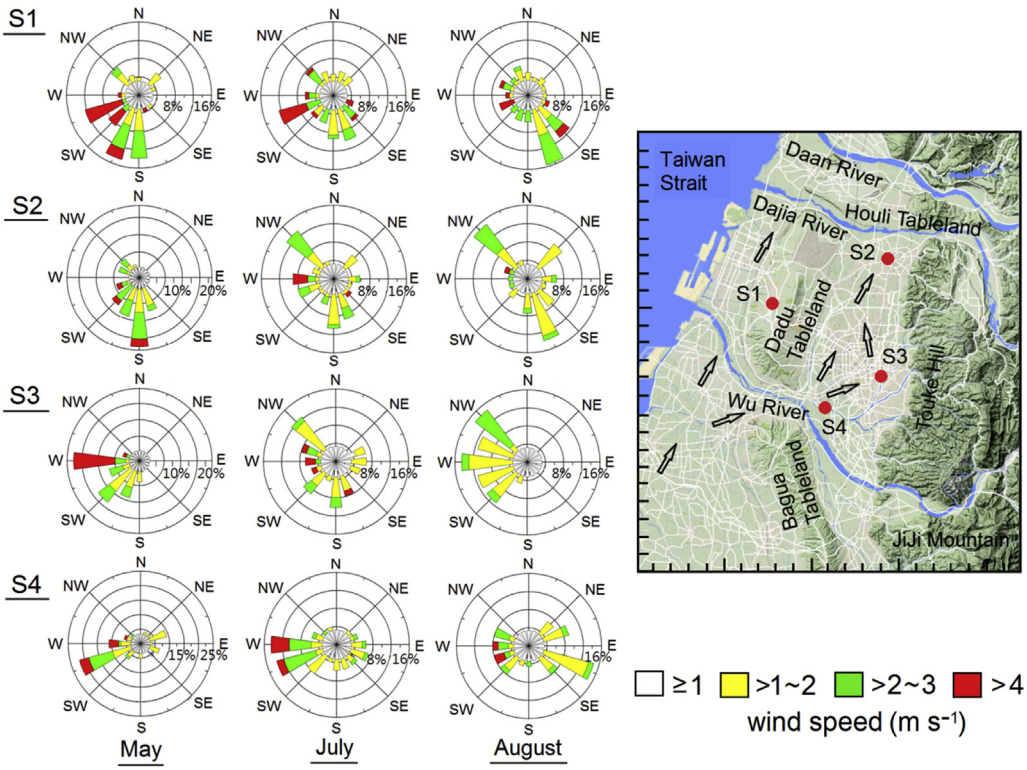

Fig. 3. Wind roses for the southwest monsoon during the sampling periods at S1–S4 sites, with simultaneously collected meteorological data for 72 h in each period to draw these wind roses.

During the southwest monsoon season, the wind blew into the basin from the pass between the Dadu and Bagua Tablelands (Fig. 3). The wind directions become more split, paralleling the spatially extensive plain trends to the north (S4 → S3 → S2). The obvious wind speed reduction and changing direction of wind were affected by the basin effect. Most of wind speeds, 90% or more, at S2–S4 were lower than 3 m s^{-1} during southwest monsoon period of the sampling, especially in August. The wind speeds at S1 were clearly higher than those at S2–S4 during the sampling period. By contrast, the wind directions at S1 were different from S2–S4. A clear monsoon effect occurred in the coastal area during the southwest monsoon.

During the northeast monsoon season (Fig. 4), the wind blew into the Taichung basin via the pass between the Dadu Tableland and Touke Hill. As mentioned above, due to the effect of open terrain on wind farms, the wind directions become more split in the basin. Same as the results during southwest monsoon, most wind speeds at S1–S4 were lower than 3 m s^{-1} in the period of the sampling. However, it is significant that the basin effect does not cause the wind speed reduction. The order of wind speeds at sampling sites was: S4 > S3 > S2 ≈ S1 during northeast monsoon period of the sampling. Therefore, contaminants were transported rapidly from north to south in the basin.

3.2. Emission source profiles of VOC determination

The major anthropogenic sources of VOCs in ambient air are the transportation sector, the use of solvents and solvent containing

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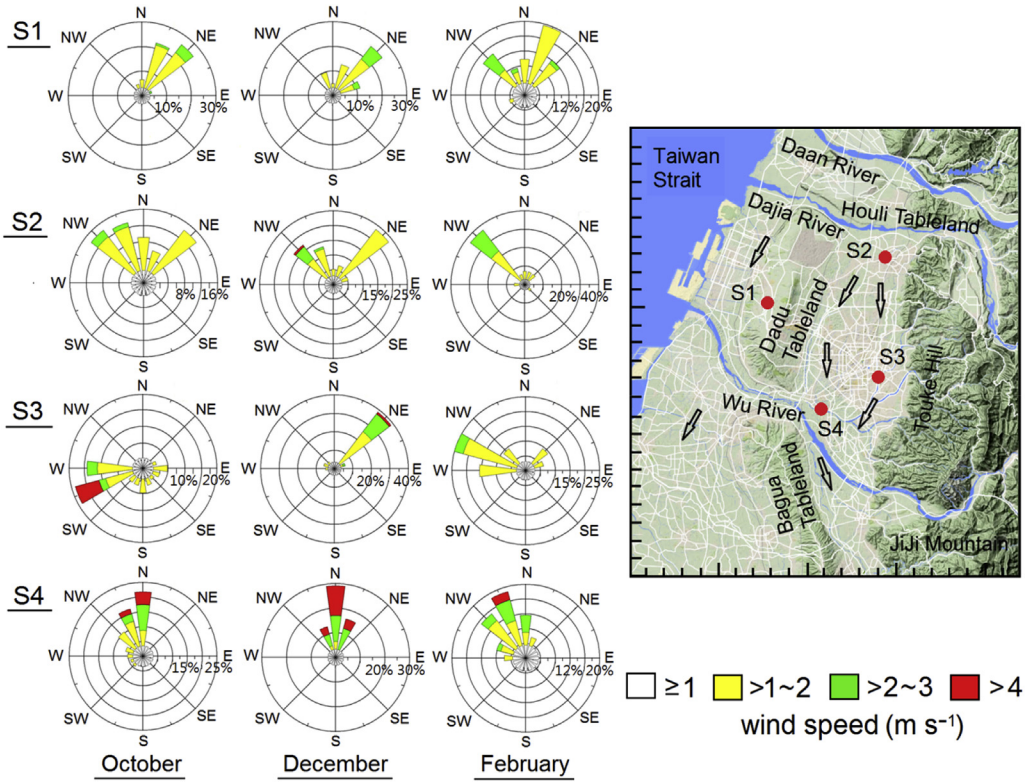

Fig. 4. Wind roses for the northeast monsoon during the sampling periods at S1–S4 sites, with simultaneously collected meteorological data for 72 h in each period to draw these wind roses.

products, and industrial sources. Vehicles are the largest source of VOC emissions. Other significant sources include petroleum production and distribution, and industrial combustion. To characterize the VOC behavior that was influenced by various emissions in this study area, the source profile of VOCs was established for each activity present in Taichung area. Ten VOCs profiles (emission from gasoline vehicle (GSE), diesel vehicle (DSE), ship (SH), gas station (GV), PVI, SCM, PR, PL, SC, and PU) were integrated from our previous studies. Because of an international commercial port is located on the west coast of Taichung area, the emission of VOCs from ships was also included in this study. The composition percentages of VOC profile of various sources in Taiwan area are listed in Table 1. The VOC profile emissions from GSE, DSE, and GV were extracted from related literature [33,34] of our laboratory. The emission data of hi-tech (including PVI and SCM) and traditional (including PR, PL, SC, and PU) industries were taken from our final study reports, which were funded by Central Taiwan Science Park Administration, National Science Council and Environmental Protection Bureau, Taichung City Government. The VOC profile emission from SH was extracted from the report by Harbor and Marine Technology Center, Taiwan [35].

3.3. VOC field measurement results

The VOC analysis results revealed 124 VOCs identified, namely 11 paraffins, 21 olefins, 7 aromatics, 25 aldehydes, 46 ketones, and 12 alcohols during southwest monsoon season. Total number of carbonyl species, aldehyde and ketone, was 71. Total concentrations of the VOCs from field measurement results at S1–S4 are shown in Fig. 5. The mean and standard deviation (mean ± SD) of total VOC concentrations at S1 (rural), S2 (suburban), S3 (urban), and S4 (suburban) were 310 ± 107, 467 ± 170, 647 ± 270, and 349 ± 108 μg m^{-3}, respectively, in May; 358 ± 203, 433 ± 280, 669 ± 235, and 434 ± 188 μg m^{-3} in July; and 337 ± 185, 260 ± 130, 570 ± 370, and 377 ± 383 μg m^{-3} in August. The average concentration of above bimestrial analysis results at S1–S4 were 335 ± 165, 387 ± 193, 629 ± 292, and 387 ± 250 μg m^{-3}, respectively. The order of average concentration and SD at sampling sites were: S3 > S4 ≈ S2 > S1 and S3 > S4 > S2 > S1, respectively, during southwest monsoon season. The transport path during the southwest monsoon entailed the wind through the pass between Dadu and Bagua Tablelands and entering the basin at S4, following the skirt of the Touke Hill to the north–northwest (S4 → S3 → S2) (Fig. 3). Most of wind speeds, 90% or more, at S2–S4 were low than 3 m s^{-1} during southwest monsoon season, especially in August. The transport of air pollutants was weak. Thus, higher VOC concentrations occurred at S3 because it located at the urban and near the main VOC emission sources (including stationary and mobile sources) (Fig. 2). By contrast, lower average concentration occurred at S1 due to its location at the rural and distant from VOC emission sources.

During northeast monsoon season, the VOC results revealed that 121 VOCs, namely 11 paraffins, 25 olefins, 7 aromatics, 28 aldehydes, 39 ketones, and 11 alcohols were identified. The average and SD of total VOCs concentrations at S1–S4 were 648 ± 226, 504 ± 271, 553 ± 153, and 425 ± 103 μg m^{-3}, respectively, in October; 798 ± 462, 835 ± 491, 715 ± 367, and 861 ± 350 μg m^{-3} in December; and 979 ± 414, 963 ± 533, 1106 ± 507, and 1026 ± 255 μg m^{-3} in February. The mean and standard deviation (mean ± SD) of total VOC concentrations at S1–S4 were: 808 ± 367, 764 ± 432, 792 ± 342, and 771 ± 236 μg m^{-3}, respectively. The average concentration and SD at S1–S4 were in the order: S1 > S3 > S4 > S2 and S2 > S1 > S3 > S4, respectively. The wind blew into the north basin from the north pass between the Dadu

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Table 1
The composition percentage of VOC profiles of various sources in Central Taiwan.

Species	Vehicle exhaust		Ship and gasoline		Hi-tech industry		Traditional industry			
	GSE	DSE	SH	GV	PVI	SCM	PR	PL	SC	PU
	$n = 28$	$n = 12$	$n = 8$	$n = 26$	$n = 30$	$n = 64$	$n = 40$	$n = 6$	$n = 7$	$n = 10$
Butane	0.7	1.4	0.0	5.0	0.0	0.1	0.6	0.1	0.0	1.8
Isobutane	0.0	7.3	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
Pentane	0.7	2.6	0.0	16.1	0.0	31.8	0.0	0.0	0.0	0.6
Isopentane	0.0	0.0	0.0	20.0	0.0	0.0	0.0	0.0	0.0	0.0
3-Methylpentane	1.0	3.2	0.0	6.6	0.0	0.0	0.0	0.0	0.0	0.0
2-Methylpentane	3.5	3.4	0.5	4.2	0.0	0.0	0.0	0.0	1.5	0.0
Hexane	2.1	3.2	0.0	1.8	0.0	0.2	0.0	0.0	3.5	0.9
2-Methylhexane	0.0	3.1	0.0	1.8	0.0	0.0	0.0	0.0	0.0	0.0
3-Methylhexane	0.0	3.2	0.3	2.0	0.0	0.0	0.0	0.0	0.0	0.0
Heptane	2.0	2.6	0.2	1.0	0.0	0.0	0.0	0.4	14.3	0.9
2-Methylheptane	0.0	1.1	0.0	1.1	0.0	0.0	0.0	0.0	0.0	0.0
3-Methylheptane	3.1	0.8	0.0	1.2	0.0	0.0	0.0	0.0	0.0	0.0
Octane	2.6	0.9	0.3	0.1	0.0	0.0	0.3	0.0	0.0	0.7
Nonane	0.2	0.1	1.0	0.1	0.0	0.0	0.0	1.6	0.0	0.3
Cyclohexane	0.0	1.3	0.0	1.6	0.0	0.0	0.0	10.7	4.6	0.0
Methylcyclohexane	0.0	0.0	0.1	0.9	0.0	0.0	0.0	0.0	7.1	0.0
Methylcyclopentane	1.9	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.8	0.0
Propene	1.2	2.6	9.1	0.2	0.0	0.7	0.0	0.8	0.0	2.6
2-Butene	1.7	1.3	3.8	1.3	0.0	1.2	1.4	0.0	0.0	2.5
1-Pentene	2.1	0.9	1.3	1.2	0.0	0.1	0.0	0.0	0.0	0.3
1-Hexene	1.8	0.0	1.3	1.4	0.0	0.7	1.7	0.0	0.0	2.1
1-Heptene	1.3	0.0	0.0	0.3	0.0	0.5	1.4	0.0	0.0	1.8
1-Octene	1.1	0.0	0.0	0.2	0.0	1.7	1.0	0.0	0.0	1.8
1-Nonene	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	1.6
Benzene	14.1	27.5	8.4	1.0	0.0	0.0	0.0	0.0	0.4	0.0
Toluene	16.1	6.2	5.2	6.9	2.6	1.9	24.9	53.7	45.1	37.9
Ethylbenzene	2.2	2.0	5.2	1.3	0.0	0.0	0.0	1.1	3.1	13.2
m,p-Xylene	5.4	4.5	3.3	4.8	0.0	0.0	0.0	3.0	0.8	0.0
o-Xylene	2.9	0.6	3.3	1.0	0.0	0.4	0.0	1.1	15.8	0.0
1,3,5-Trimethylbenzene	1.3	0.2	0.8	1.1	0.0	0.3	0.0	2.1	0.0	0.0
1,2,4-Trimethylbenzene	0.0	0.1	3.2	0.8	0.0	0.0	0.0	1.8	0.0	0.0
Acetaldehyde	5.3	0.0	0.0	0.0	11.1	4.3	7.2	0.0	0.0	3.3
Butyraldehyde	4.1	0.0	0.0	0.0	8.4	2.1	4.0	0.0	0.0	1.8
Valeraldehyde	3.6	0.0	0.0	0.0	19.4	4.7	4.8	0.0	0.0	4.4
Isovaleraldehyde	0.8	0.0	0.0	0.0	0.0	0.1	1.5	0.8	0.0	0.7
Caproaldehyde	3.4	0.0	0.0	0.4	0.0	3.6	0.0	0.0	0.0	2.9
Acetone	4.3	0.0	7.5	0.0	5.1	0.0	0.0	3.7	0.0	1.6
Ethylmethylketone	3.2	0.0	2.6	0.0	9.8	0.1	18.4	3.7	0.0	4.5
2-Pentanone	0.0	0.0	0.0	0.0	8.0	10.6	4.0	0.0	0.0	0.0
Isopropylmethylketone	0.7	0.0	0.0	0.0	0.0	0.2	1.0	0.0	0.0	0.6
MethylIsobutylKetone	0.4	0.0	0.1	0.0	0.0	0.7	1.1	0.0	0.0	0.4
Cyclohexanone	0.0	0.0	0.0	0.0	0.0	0.0	0.6	1.7	0.0	5.3
Ethanol	1.2	0.0	0.0	0.2	3.7	0.7	1.8	0.0	0.0	0.0
1-Butanol	2.3	8.0	0.0	0.0	20.6	9.8	4.2	6.8	0.0	1.2

GSE gasoline engine, *DSE* diesel engine, *SH* ship, *GV* gasoline station, *PVI* photovoltaic industry, *SCM* semiconductor manufacturing, *PR* printers, *PL* plastics, *SC* surface coating, *PU* polyurethane synthetic leather, n = the number of measurement.

Tableland and Touke Hill and subsequently skirted along the east of the Touke Hill (S2 → S3 → S4). In the basin, the wind direction varied because of the blocking effect of Touke Hill, which was the same situation during the southwest monsoon season but with the opposite direction. Similar to the southwest monsoon season, the mean wind speed at S2–S4 ranged from 1 to 3 m s^{-1}. Therefore, the transport of air pollutants was also weak. Highest average concentration of VOCs occurred at S1, which may result from the local emission sources with lower wind speed (<2 m s^{-1}) (Fig. 3b).

Total spices number of VOCs and carbonyls during the northeast monsoon season were slightly lower than that those during the southwest monsoon season (northeast monsoon 124 VOCs, including 71 carbonyls; southwest monsoon 121 VOCs, including 67 carbonyls).

Comparison the total concentrations during both monsoon seasons, the ranges of average ratio (northeast to southwest monsoon seasons) of total VOC concentrations and standard deviations at S1–S4 were 2.41, 1.98, 1.26, and 1.99 and 2.22, 2.24, 1.17, and 0.94, respectively. The difference may be due to the mixing height; during northeast monsoon season it is obviously lower than southwest one. Furthermore, the obtained results during the both monsoon seasons demonstrate that the effect of urban heat island phenomena on the area was negligible, especially during the southwest monsoon season.

3.4. Carbonyl concentrations

As mentioned previously, the present study employed the US EPA TO-15 Method to measure ambient carbonyl and VOC concentrations. The air sample was collected and stored in the specially prepared (without humidification) silcosteel canisters, and then analyzed by GC/MS with a preconcentrator. Trace oxygen after preconcentrator may interfere sample qualitative analysis. The mass range of the electron impact ionization mass spectrometer was selected from 33 to 300 atomic mass units (amu) in this study to avoid this interference, in which formaldehyde was not measured due to its molecular mass lower than 33 amu. Lu et al. [36] reported that the concentration ratios of formaldehyde/

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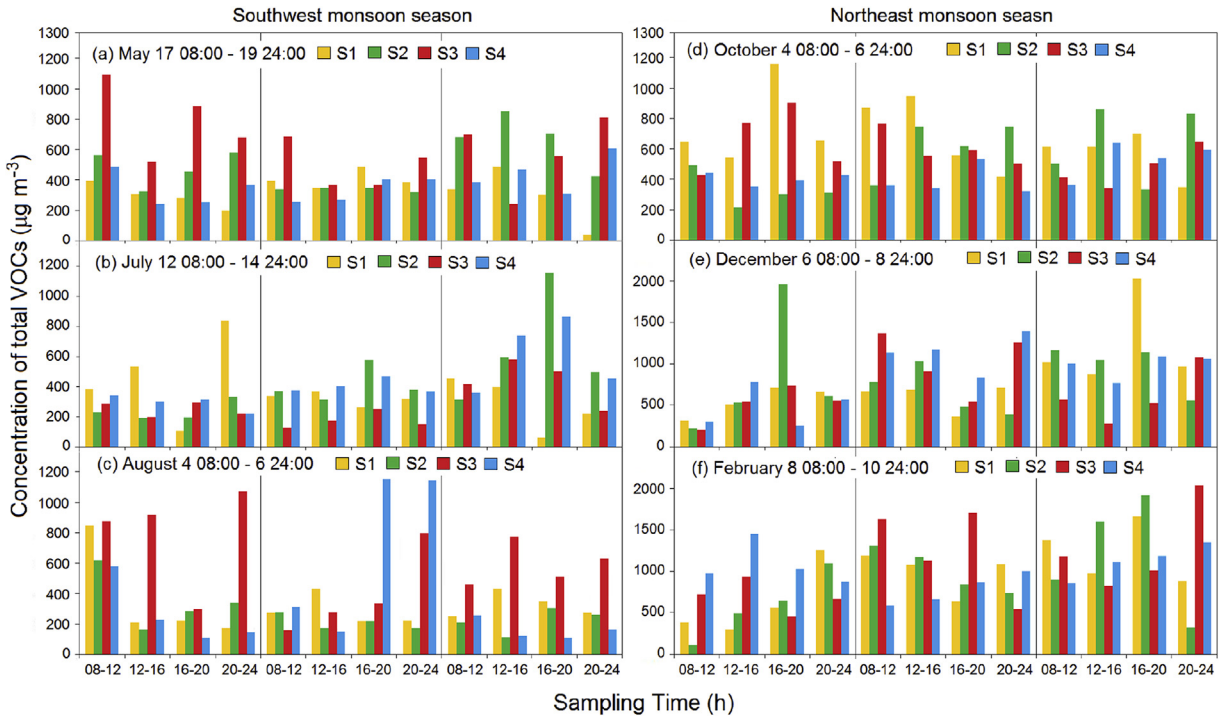

Fig. 5. Total concentrations of the VOC field measurement results at sampling sites S1–4. Note that the scales of concentration of total VOCs in (e) and (f) are different from (a)–(d). Notice that the scale of total VOC concentration in (e) and (f) are difference from others.

acetaldehyde varied from 0.69 to 1.06 with a mean of 0.84 in the clear days and from 0.59 to 1.24 with a mean of 0.87 in the hazy days in Guangzhou, China. These ratios were quite similar to those reported in the three urban areas of the United States (mean for all data, 1.10) [37] and in the urban environments of Hong Kong and Beijing (normally vary from 1 to 2) [38–40]. Thus, these results might be used to estimate the concentration of ambient formaldehyde.

A total of 71 and 67 carbonyls (aldehydes and ketones) were analyzed from sites S1–S4 during the southwest and northeast monsoon periods, respectively in this study. The concentrations of several carbonyl compounds were too low (<5 μg m^{-3}) to be discussed, such as decanal, undecanal, 2-decanone, 2-undecanone, 2,3-butanedione, 2,3-pentanedione, 2,3-hexanedione, 2,5-hexanedione, 2,7-octanedione, 2,3-octanedione, isopropyl vinyl ketone, allyl methyl ketone, crotonaldehyde, and 8-nonen-2-one. Moreover, in order to simplify the description of the phenomena, carbonyl compounds with n carbon atoms (including straight-chain, branched, cyclic, and carbon–carbon double bond structures) were integrated into total C_n aldehydes ($\sum C_n$-al) or total C_n ketones ($\sum C_n$-one) in this study. For example, $\sum C_4$-one means the sum of ethyl methyl ketone and 3-buten-2-one concentrations. The main aldehydes and ketones from S1–S4 ambient air samples are summarized in Table 2.

Fig. 6 represents the box-whisker plots of the results of carbonyl concentrations at sites S1–S4 in Taichung area during the southwest monsoon season. The averages of total aldehyde and ketone concentrations at S1 were 60 and 68 (morning), 53 and 59 (afternoon), 34 and 36 (evening), and 39 and 49 (night) μg m^{-3}; at S2 were 46 and 72 (morning), 40 and 54 (afternoon), 53 and 74 (evening), and 42 and 60 (night) μg m^{-3}; at S3 were 95 and 96 (morning), 69 and 78 (afternoon), 53 and 78 (evening), and 70 and 88 (night) μg m^{-3}; and at S4 were 49 and 60 (morning), 40 and 47 (afternoon), 55 and 63 (evening), and 54 and 62 (night) μg m^{-3}, respectively. From these results, it can be summarized that: (a) the major carbonyls in ambient were ketones, (b) the concentrations of carbonyls at morning, afternoon, evening, and night were in the order: S3 > S1 > S2 > S4, S3 > S1 > S2 > S4, S3 > S2 > S4 > S1, and S3 > S4 > S2 > S1, respectively, (c) higher carbonyl concentrations occurred at S3 (urban) all day, (d) VOC concentration at downwind (S2) was higher than at upwind (S4) except during the night, and (e) the maximum variation carbonyl concentrations occurred at S1 (rural) with high and low concentration at daytime and nighttime, respectively.

The box-whisker plots of the analysis results of carbonyl concentrations at sites S1–S4 in Taichung area during the northeast monsoon season are shown in Fig. 7. The average of total aldehyde and ketone concentrations at S1 were 95 and 115 (morning), 87 and 118 (afternoon), 118 and 127 (evening), and 94 and 121 (night) μg m^{-3}; at S2 were 79 and 98 (morning), 101.9 and 126.9 (afternoon), 81.38 and 103.5 (evening), and 82.7 and 102.0 (night) μg m^{-3}; at S3 were 84.4 and 104.5 (morning), 81.8 and 103.8 (afternoon), 94.4 and 118.8 (evening), and 78.8 and 99.3 (night) μg m^{-3}; and at S4 were 87.7 and 110.5 (morning), 95.5 and 118.4 (afternoon), 94.3 and 121.1 (evening), and 100.3 and 132.1 (night) μg m^{-3}, respectively. From the results, it can be summarized that: (a) same as during the southwest monsoon season, the major carbonyls in ambient were ketones, (b) the concentrations of carbonyls at morning, afternoon, evening, and night were in the order: S1 > S4 > S3 > S2, S2 > S4 > S1 > S3, S1 > S4 ≈ S3 > S2, and S4 > S1 > S2 ≈ S3, respectively, (c) the concentrations of carbonyl at all day were: S1 (rural) > S4 (suburban) ≈ S2 (suburban) > S3 (urban), which was difference from during the southwest monsoon season, (d) VOCs concentration corresponding to the wind direction was weak (S4 ≈ S2), (e) the variations of carbonyl concentration at S1–S4 were all low, and (f) the ranges of average concentration ratio (northeast to southwest monsoon seasons) of total carbonyls at S1–S4 were 1.6–3.5 with mean of 2.4, 1.5–2.5 with mean of 1.8, 1.0–1.6 with mean of 1.3, and 1.8–2.4 with mean of 2.0, respectively.

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Table 2
Summary of main aldehydes and ketones from S1–S4 ambient air samples.

Number of carbon	Carbonyl compounds			
Aldehydes				
C_2	Acetaldehyde			
C_3	2-Propenal			
C_4	n-Butyraldehyde	Isobutyraldehyde	2-Methyl-2-propenal	
C_5	2-Pentenal	4-Pentenal	Valeraldehyde	2-Methylbutyraldehyde
	Isovaleraldehyde			
C_6	4-Methyl-3-pentenal	2-Methylvaleraldehyde	3-Methylvaleraldehyde	n-Caproaldehyde
	Benzaldehyde	2-Hexenal		
C_7	n-Heptanal	2-Methylhexanal	3-Metthylhexanal	4-Methylhexanal
	5-Methylhexanal	2,3-Dimethylpentanal		
C_8	Octanal	2-Methylheptanal		
C_9	n-Nonanal			
Ketones				
C_3	Acetone			
C_4	Ethyl methyl ketone	3-Buten-2-one		
C_5	2-Pentanone	3-Pentanone	1-Penten-3-one	Isopropyl methyl ketone
	3-Penten-2-one	Cyclopentanone		
C_6	Methyl Isobutyl Ketone	4-Hexen-2-one	5-Hexen-2-one	Butyl methyl ketone
	Ethyl propyl ketone	Ethyl isopropyl ketone	2-Methylcyclopentanone	3-Methylcyclopentanone
	Cyclohexanone	2-Methyl-3-pentanone	4-Methyl-4-penten-2-one	3-Methyl-2-pentanone
C_7	4-Heptanone	Butyl ethyl ketone	4-Methyl-2-hexanone	5-Methyl-2-hexanone
	5-Methyl-3-hexen-2-one	5-Methyl-5-hexen-2-one	Isopropyl propyl ketone	3-Methylcyclohexanone
	Amyl methyl ketone			
C_8	2-Octanone	3-Octanone	4-Methyl-2-heptanone	5-Methyl-2-heptanone
	6-Methyl-2-heptanone	3-Methylcyclohexanone	2,3-Dimethylcyclohexanone	3,4-Dimethyl-2-hexanone
C_9	8-Nonen-2-one	2-Nonanone	4,6-Dimethyl-2-heptanone	

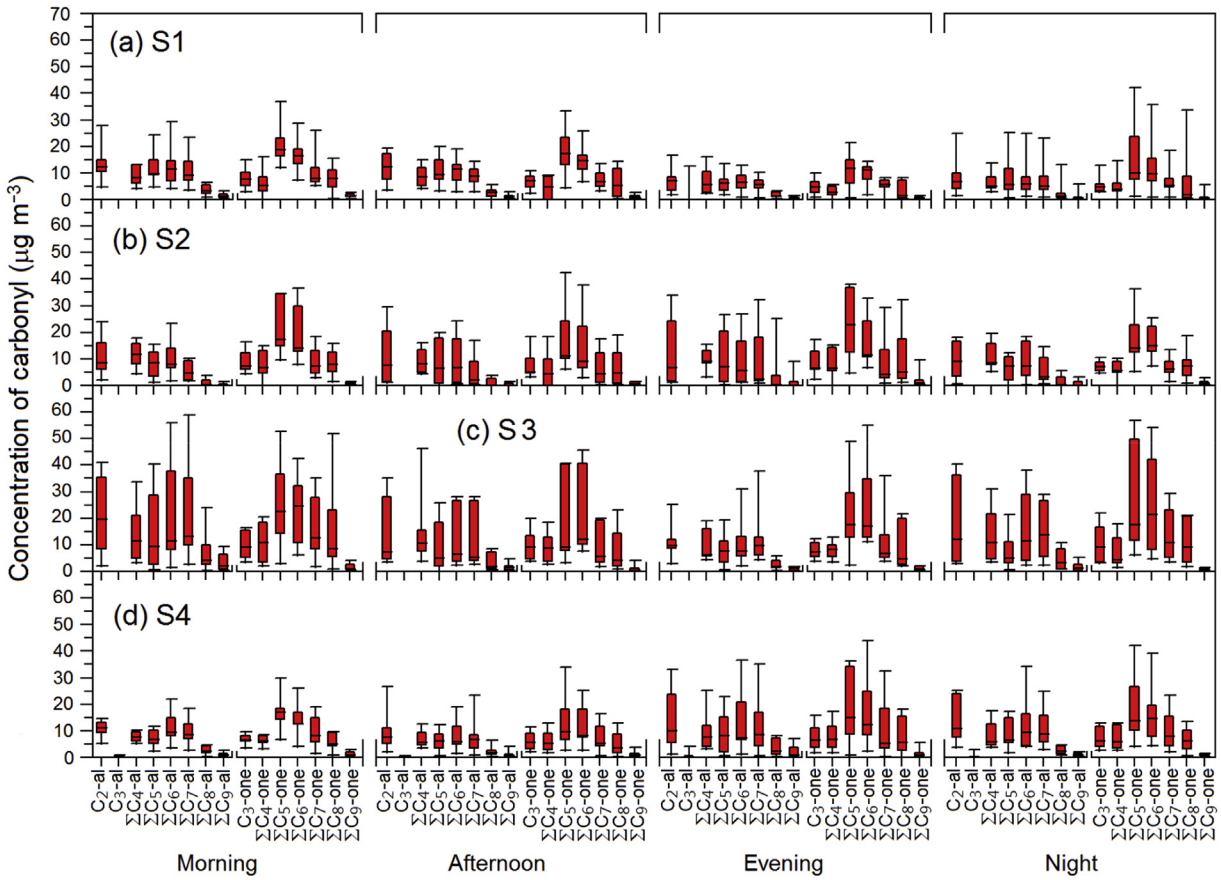

Fig. 6. Box-whisker plots of the analysis results of carbonyls at four sampling sites in Taichung area during the southwest monsoon season.

3.5. Simulation results of VOCs by CMB8.2 model

Because in-situ atmospheric formation is a major source of carbonyls [41], therefore, the simulation results of VOCs sources by CMB8.2 receptor model can be used to understand the primary sources of ambient carbonyls. Figs. 8 and 9 show the box-whisker plots of source contribution of VOCs at the four sampling sites in Taichung area during southwest and northeast

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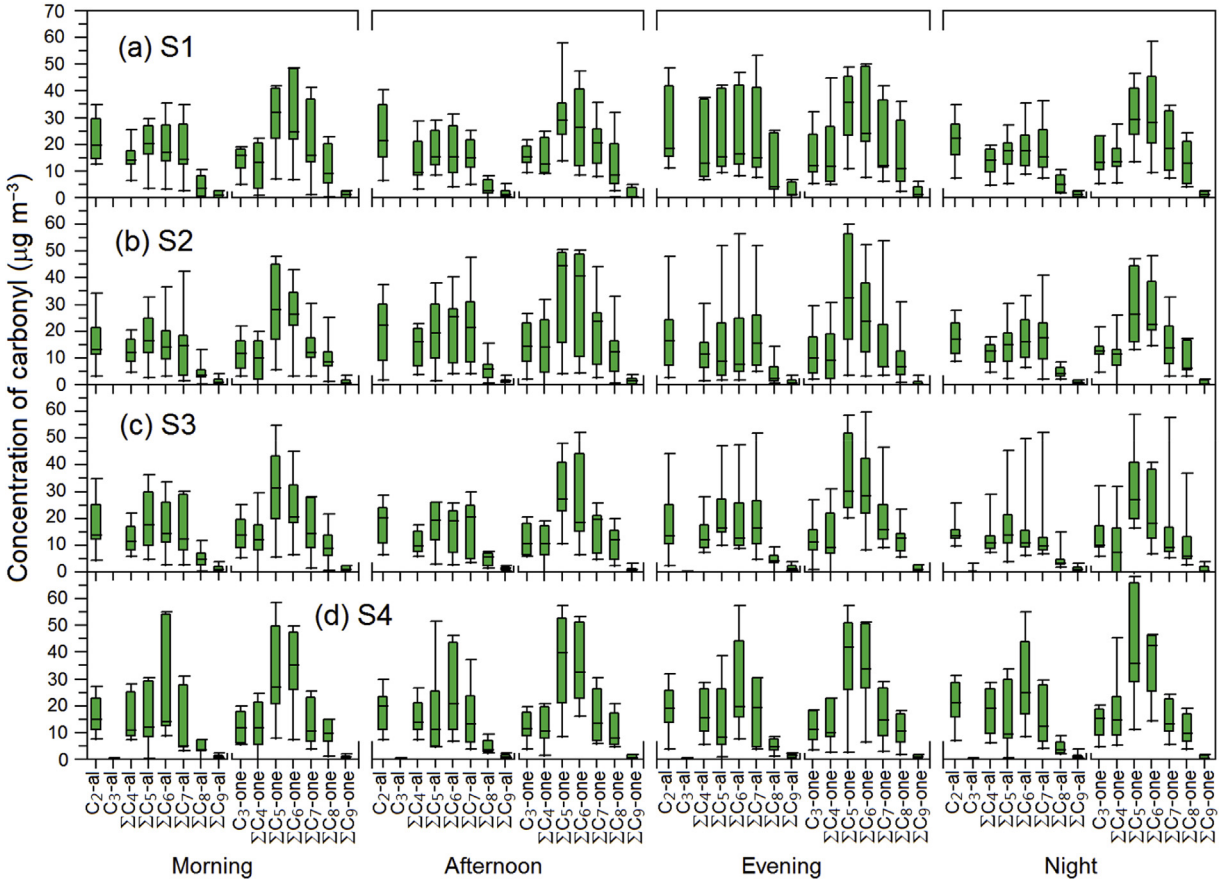

Fig. 7. Box-whisker plots of the analysis results of carbonyls at four sampling sites in Taichung area in the northeast monsoon season.

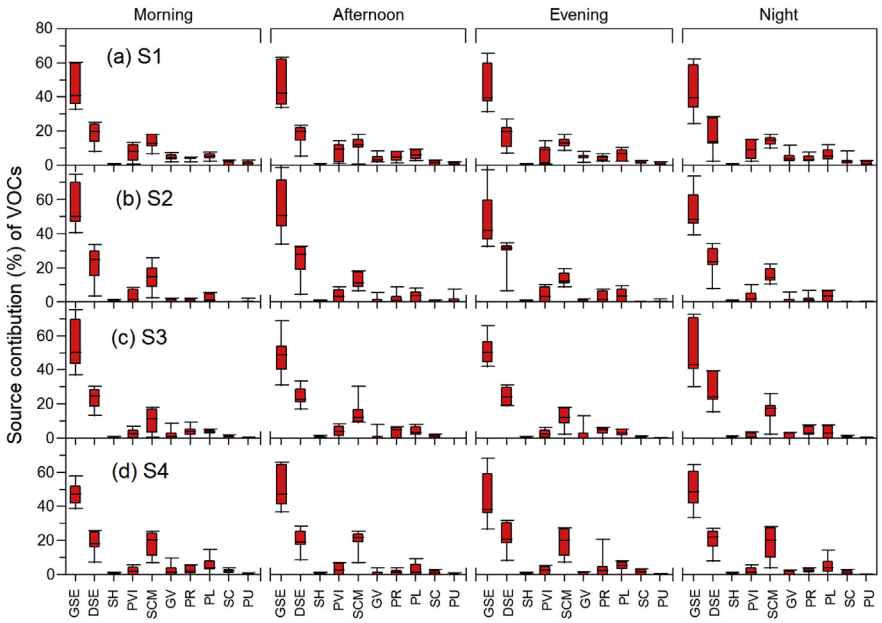

Fig. 8. Box-whisker plots of sources contribution of VOCs to the four sampling sites in Taichung area from southwest monsoon season.

monsoon seasons, respectively. As mentioned above, the performance of the CMB8.2 model was evaluated by using three statistical indices: $R^2 \geq 0.8$, $\chi^2 \leq 4.0$, and % mass = 80–120%. The R^2, χ^2, and % mass values during the southwest and northeast monsoon seasons varied from 0.862–0.965, 1.6–2.1, and 91–104% and 0.893–0.971, 1.3–2.0, and 93–99%, respectively; therefore, the obtained source contributions for the VOCs were relatively accurate.

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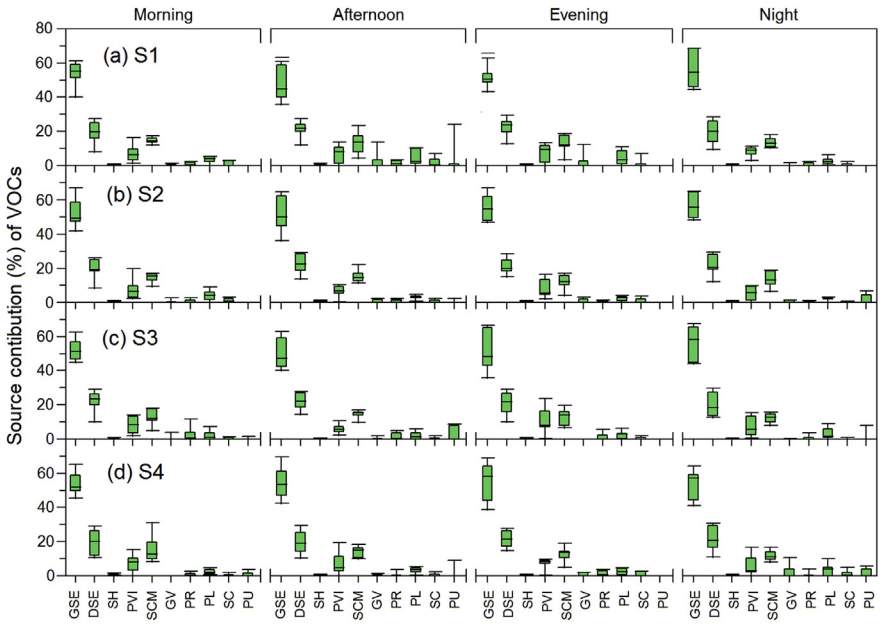

Fig. 9. Box-whisker plots of sources contribution of VOCs to the four sampling sites in Taichung area from northeast monsoon season.

From Fig. 8, it can be found that the contributions of VOCs sources were in the order: vehicle exhaust (GSE and DSE) > hi-tech industry (PVI and SCM) > traditional industry (PR, PL, SC, and PU) > ship and gasoline stations (SH and GV) during the southwest monsoon season. The contribution ranges of VOCs from vehicle exhaust at S1–S4 were 58–63, 75–78, 72–77, and 66–70%, respectively. Hi-tech industry (PVI and SCM) contributed from 19–24, 16–18, 13–18, and 2–23% to sites S1, S2, S3, and S4, respectively. Taichung Science Park is located on the east foothill of Dadu Tableland, and by the end of 2012, there were 161 factories, 8 incubation centers, and 6 research units in the park. Because of S1 proximity to this park, the contribution of VOCs from hi-tech industry to S1 was higher than to other sites.

During the northeast monsoon season, the order of contributions of VOCs sources were vehicle exhaust (GSE and DSE) > hi-tech industry (PVI and SCM) > traditional industry (PR, PL, SC, and PU) > ship and gasoline stations (SH and GV) (Fig. 9), which was the same as during the southwest monsoon season. Vehicle exhaust (GSE and DSE) was major VOC sources during the northeast monsoon season, contributing about 68–74, 72–77, 73–75, and 73–75% to sites S1, S2, S3, and S4, respectively; with highest contribution occurring at night. The contribution ranges of VOCs from hi-tech industry at S1, S2, S3, and S4 were 19–22, 18–21, 19–22, and 17–22%, respectively, with relative higher contribution at S1 and S3 (average 21%).

The concentrations of ambient carbonyls increase clearly with increasing total VOC concentrations (Figs. 5–7). As mentioned above, motor vehicles are the leading source of carbonyls emission in non-industrial area. A total of 59 carbonyls were quantified from the exhausts of gasoline-powered light-duty (LDVs) and heavy-duty diesel-powered (HDVs) vehicles [11]. And photochemical reactions made significant contributions to the ambient levels of carbonyls in summer whereas in winter motor vehicle emissions would be the major sources of the carbonyls [42]. Therefore, in order to effectively reduce the carbonyls in ambient air, pollution from vehicle exhaust must be managed and controlled.

4. Conclusions

In this study, a specially prepared (without humidification) silcosteel canisters with air sampling kites was used for collecting ambient air. It successfully avoids unstable state of unsaturated carbonyls in canisters. A total of 71 and 67 carbonyls (aldehydes and ketones) from four sampling sites were determined by GC/MS during the southwest and northeast monsoon periods, respectively. The total aldehyde concentrations were lower than total ketone concentrations during the southwest and northeast monsoon seasons. The concentrations of carbonyls at both daytime and nighttime were in the order: urban > rural > suburban and urban > suburban > rural areas. Higher carbonyl concentrations occurred in the urban area, but the maximum variation of carbonyl concentrations occurred in the rural area during the southwest monsoon season. By contrast, the order of carbonyl concentrations at daytime and nighttime was irregular during the northeast monsoon season; the variations of carbonyl concentrations at urban, suburban, and rural were all low. The ranges of average concentration ratio (northeast to southwest monsoon seasons) of total carbonyls in urban, suburban, rural areas were 1.0–1.6, 1.5–3.5 or 1.5–2.5, and 1.8–2.4, respectively.

The results demonstrated that during the both monsoons, the effect of urban heat island phenomena on the area was negligible, especially during the northeast monsoon season. Taichung basin had already been overdeveloped and the transport of air pollutants in the basin was weak. However, the photochemical reactions in the area were evident, so the total spices number of carbonyls was very large during the both monsoon seasons.

The results during the both monsoon seasons indicated that the concentrations of ambient carbonyls increased with increasing total VOC concentrations. The use of simulation results of VOCs by CMB8.2 model to understand the contribution from primary sources of carbonyls is useful. GSE, DSE, PVI, and SCM were four major sources of VOCs and carbonyls during the both monsoon seasons. Therefore, in order to effectively reduce the carbonyls in ambient air, pollution from vehicle exhaust and hi-tech industry must be managed and controlled.

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